

Universal thermodynamic limit of quantum engine efficiency

Wolfgang Niedenzu,^{1,*} Victor Mukherjee,¹ Arnab Ghosh,¹ Abraham G. Kofman,^{1,2} and Gershon Kurizki¹

¹*Department of Chemical Physics, Weizmann Institute of Science, Rehovot 7610001, Israel*

²*CEMS, RIKEN, Saitama, 351-0198, Japan*

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The efficiency of heat engines is limited by the Carnot bound, attained when the engine cycle is reversible. Quantum engines fuelled by non-thermal (e.g., squeezed-thermal) baths have been shown to surpass this bound. Yet, their maximum efficiency cannot be determined by the reversibility condition, which may yield an unachievable efficiency bound above 1. This prompts the question: What does really limit the efficiency? We identify the fraction of the exchanged energy between a quantum system and a bath that necessarily causes an entropy change and derive a new inequality for the latter. This formulation reveals a universal efficiency bound for quantum engines which is invariably attained for the least dissipation over the engine cycle but does not imply reversibility, unless the baths are thermal. This bound thus cannot be solely deduced from the laws of thermodynamics. We illustrate these results for the practically-relevant Carnot- and Otto cycles energised by non-thermal baths, which are both shown to be restricted by our new efficiency bound.

I. INTRODUCTION

Engines are machines that convert some form of energy (e.g., thermal or electrical energy) into work. Their efficiency, defined as the ratio of the extracted work to the invested energy, is restricted to 1 at most by the energy-conservation law. While mechanical engines may reach this bound, Carnot showed [1] that the efficiency of any heat engine that cyclically operates between two thermal baths is universally limited by the ratio of the bath temperatures, regardless of the concrete design [2, 3]. The universality of this bound led to the formalisation of the second law of thermodynamics and the introduction of the notion of entropy by Clausius [4]. In terms of entropy, the Carnot bound is attained by (idealised) heat engines that operate reversibly, so that the total entropy of the engine and the baths is unaltered over a cycle [2, 3]. This corresponds to the minimum amount of heat being dumped into the cold bath, so as to close the cycle, and hence to the maximum input heat being transformed into work. By contrast, in an irreversible cycle, a larger amount of heat must be dumped into the cold bath, so that less input heat is available for conversion into work, causing the engine efficiency to decrease [3, 4].

As part of the effort to understand the rapport between quantum mechanics and thermodynamics [5–19] (see [20–23] for recent reviews), the Carnot bound has been challenged for quantum engines that exploit non-thermal quantum baths [9, 24–30]. In this respect, a distinction is to be drawn between two types of non-thermal engines [29, 31], (i) engines wherein the working medium equilibrates to a thermal state whose temperature is adjustable (e.g., by the phase of the coherence in a “phaseonium” bath [9]), which qualify as genuine heat engines with a controllable Carnot bound, and (ii) engines wherein the non-thermal (e.g., squeezed [27]) bath may

render the working-medium state non-thermal, making the Carnot bound irrelevant.

The efficiency bound of the latter type of engines has been addressed [26, 27, 29, 32] but still needs elucidation: What is particularly puzzling is that, contrary to heat engines, their efficiency bound cannot be deduced from the requirement of reversible operation: Reversibility may entail an efficiency bound that not only surpasses the Carnot bound but also unity [30], making it unachievable. Hence, the question naturally arises whether such engines are limited by constraints other than the second law.

The second law for quantum relaxation processes is widely accepted [8, 17, 20–23, 30, 33–42] to be faithfully rendered by Spohn’s inequality [43]. Here we show that the inadequacy of assessing the maximum efficiency via this standard reversibility criterion arises whenever the engine’s working medium is in a non-passive quantum state [6, 7, 10, 21–23, 29, 31, 40, 44–53]. The definition [6, 7, 10] of a non-passive state is that its energy can be *unitarily* reduced until the state becomes passive, thereby extracting work. Non-passive states may thus be thought of as being “quantum batteries” [45, 48] or “quantum flywheels” [54] (Fig. 1). The maximum amount of work extractable from such states (their “work capacity”) has been dubbed “ergotropy” in Ref. [10]. For example, every population-inverted state is non-passive and so are, e.g., coherent or squeezed field states, whereas thermal states are passive.

As we show, the standard reversibility criterion provides an inequality for the change in the engine entropy which may be much too loose (non-tight) to be useful if non-passive states are involved. The distinction between non-passive and passive states is at the heart of our analysis and underlies our division of the energy exchanged between a quantum system and a bath into a part that *necessarily* causes an entropy change, and ergotropy. Our proposed division is in fact a new unraveling of the first law of thermodynamics for quantum systems.

In scenarios where non-thermal baths may create non-passive states of the working medium, we derive a new

* Wolfgang.Niedenzu@weizmann.ac.il

inequality for the entropy change which yields a physical efficiency limit of the engine that never surpasses unity. This efficiency limit is universal and in general cannot be assessed by the standard reversibility criterion. Concrete examples of such engines are given.

II. THE FIRST LAW OF QUANTUM THERMODYNAMICS

For an arbitrary process taking the initial state ρ_0 of a quantum system to an evolving state $\rho(t)$, which may be governed by a time-dependent Hamiltonian $H(t)$ and a bath, energy conservation implies

$$\Delta E(t) = \mathcal{E}_d(t) + W(t), \quad (1)$$

where $\Delta E(t)$ is the change in the system energy $E(t) = \text{Tr}[\rho(t)H(t)]$. Its two constituents are

$$\mathcal{E}_d(t) := \int_0^t \text{Tr}[\dot{\rho}(t')H(t')]dt', \quad (2a)$$

which is the non-unitary dissipative energy change due to the interaction with the bath, and

$$W(t) := \int_0^t \text{Tr}[\rho(t')\dot{H}(t')]dt', \quad (2b)$$

which is the work [6] due to changes of the system Hamiltonian. Contrary to the energy change $\Delta E(t)$, both $\mathcal{E}_d(t)$ and $W(t)$ are process variables that generally depend on the evolution path, not only on the initial and final states. For thermal baths, the energy $\mathcal{E}_d(t)$ is commonly identified with the transferred heat [8]. The energy $\mathcal{E}_d(t)$ vanishes for a closed (isolated) system whose state evolves unitarily according to the von Neumann equation $\dot{\rho}(t) = \frac{1}{i\hbar}[H(t), \rho(t)]$. If the Hamiltonian is constant, then there is no work, $W(t) = 0$.

We here consider general scenarios, wherein the bath and/or the system may be in a non-thermal state and strive to better understand the nature of the exchanged energy (2a) and, in particular, its relation to entropy change. As we show, only part of the exchanged energy $\mathcal{E}_d(t)$ is necessarily accompanied by a change in entropy.

To elucidate this issue, we resort to the concept of non-passive states (see Fig. 1 and Appendix A). The energy $E(t)$ of a non-passive state $\rho(t)$ can be decomposed into ergotropy $\mathcal{W}(t) \geq 0$ and passive energy $E_{\text{pas}}(t)$. Ergotropy is the maximum amount of work that can be extracted from such a state by means of unitary transformations [6, 7, 10]. By contrast, the passive energy, which is the energy of the passive state $\pi(t)$, cannot be extracted in the form of work.

The von Neumann entropy $\mathcal{S}(\rho(t)) = -k_B \text{Tr}[\rho(t) \ln \rho(t)]$ of a non-passive state $\rho(t)$ is the same as that of its passive state $\pi(t)$ since the two are related by a unitary transformation. Hence, a change in entropy requires a change in the passive state

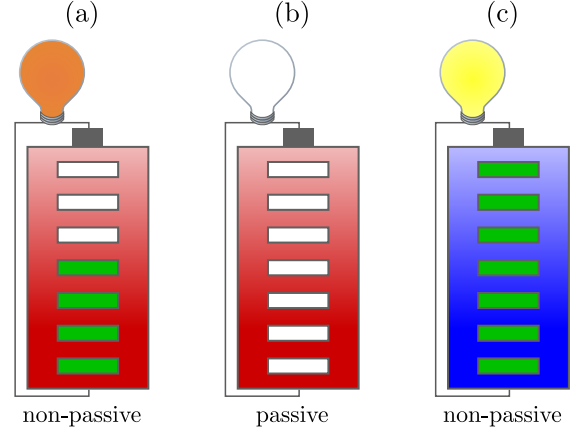


Figure 1. The different kinds of energy contained in a quantum state visualised by means of a battery at a certain temperature. The battery charge (green bars) represents ergotropy \mathcal{W} (extractable as work, here illustrated by a lighted bulb) and its temperature (colour of the battery: red—hot, blue—cold) represents passive (here: thermal) energy E_{pas} —the higher the temperature the larger the passive energy. (a) The battery is partly charged and hot: This represents a non-passive state which allows for work extraction. As the battery is not completely charged, the light bulb appears dim. (b) The battery is discharged, but its temperature is the same as in (a). This state is the passive state of (a) and, consequently, the light bulb does not shine. (c) The battery is in a non-passive state whose ergotropy is higher than in (a) (the battery is fully charged) but the passive energy is lower (the battery is colder). Although the total energy in (a) and (c) may be the same, more work can be extracted from the state (c), causing the light bulb to shine brighter than in (a).

$\pi(t)$. Equation (2a), however, does not discriminate between $\rho(t)$ and $\pi(t)$: A change in $\rho(t)$ may cause a non-zero $\mathcal{E}_d(t)$ but not necessarily a change in entropy. By contrast, a change in $\pi(t)$ results in entropy change.

In order to explicitly account for a change in the *passive* state, we may decompose the dissipative energy change (2a) as follows,

$$\mathcal{E}_d(t) = \Delta E_{\text{pas}|d}(t) + \Delta \mathcal{W}|_d(t), \quad (3)$$

where

$$\Delta E_{\text{pas}|d}(t) := \int_0^t \text{Tr}[\dot{\pi}(t')H(t')]dt' \quad (4a)$$

is the dissipative (non-unitary) change in passive energy and

$$\Delta \mathcal{W}|_d(t) := \int_0^t \text{Tr}[(\dot{\rho}(t') - \dot{\pi}(t'))H(t')]dt' \quad (4b)$$

is the dissipative (non-unitary) change in the system ergotropy due to its interaction with the bath. The microscopic decomposition of the exchanged energy (3) into dissipative change in passive energy (4a) and dissipative ergotropy change (4b) is a new unraveling of the first law

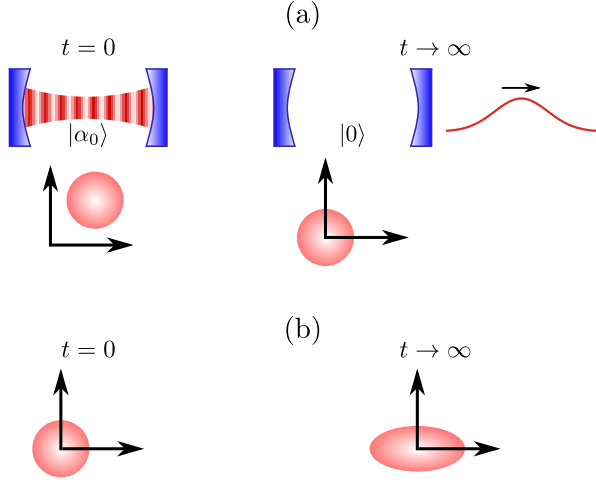


Figure 2. (a) A cavity mode initialised in a coherent state decays into the surrounding electromagnetic-field bath to the vacuum state. (b) A cavity mode prepared in the vacuum state evolves to a squeezed-vacuum state due to its interaction with a squeezed bath. The circles and the ellipse represent the respective phase-space distributions [56] of the field states.

of thermodynamics for quantum systems that constitutes one of our main results.

The decomposition (3) carries with it the following insights: (a) Although ergotropy may be transferred from a non-thermal bath to the system in a non-unitary fashion, it may afterwards still be extracted from the system in the form of work via a suitable unitary transformation. (b) Consistently, any *unitary* changes (in either ergotropy or in passive energy due to time-dependent changes of the Hamiltonian) are associated with work (2b). If the Hamiltonian is constant, then $\Delta E_{\text{pas}}|_{\text{d}}(t)$ is only the change in passive energy without work, $\Delta E_{\text{pas}}|_{\text{d}}(t) = \Delta E_{\text{pas}}(t) = \text{Tr}[\pi(t)H] - \text{Tr}[\pi_0 H]$, where π_0 is the passive counterpart of the initial state ρ_0 . Likewise, $\Delta \mathcal{W}|_{\text{d}}(t) = \Delta \mathcal{W}(t) = \mathcal{W}(\rho(t)) - \mathcal{W}(\rho_0)$ is then the change in ergotropy without work performance. (c) While a non-zero $\Delta E_{\text{pas}}|_{\text{d}}(t)$ entails a change in the passive state $\pi(t)$ and hence in entropy, a non-zero $\mathcal{E}_{\text{d}}(t)$, by contrast, does not necessarily imply an entropy change, as shown below. The correspondence of $\Delta E_{\text{pas}}|_{\text{d}}(t)$ and $\Delta \mathcal{S}(t)$ is plausible since they have the *same* sign provided a majorisation relation [40, 55] holds for $\rho(t)$, as detailed in Appendix B.

Let us illustrate these insights for a single cavity mode (harmonic oscillator at frequency ω) prepared in a pure coherent state $\rho_0 = |\alpha_0\rangle\langle\alpha_0|$ that interacts (via a leaky mirror) with the surrounding electromagnetic-field bath (Fig. 2a), which for optical frequencies is very close to the vacuum state [56]. Being in contact with a bath, the cavity-mode state evolves in a non-unitary fashion (according to a quantum master equation [42]). Since the Hamiltonian is constant, the work (2b) vanishes, $W(t) = 0$. While the cavity field exponentially decays to the vacuum state, $\rho(t) = |\alpha_0 e^{-\kappa t}\rangle\langle\alpha_0 e^{-\kappa t}|$, where κ is

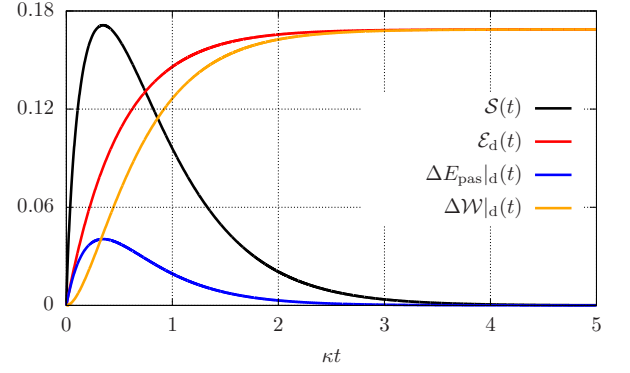


Figure 3. Entropy and energy changes for a single cavity mode prepared in the vacuum state that interacts with an outside bath in a squeezed-vacuum state (Fig. 2b) obtained by a numerical integration of the master equation. The energies are given in units of $\hbar\omega$ and the entropy in units of k_{B} . Parameters: $\omega = 10\kappa$ and squeezing parameter $r = 0.4$, κ being the decay rate of the cavity.

the leakage rate, its entropy does not change, $\mathcal{S}(\rho(t)) = 0$, so that the passive state $\pi(t) = |0\rangle\langle 0|$ is constant. Consequently, $\Delta E_{\text{pas}}|_{\text{d}}(t) = 0$ and the entire energy change is due to dissipated ergotropy, $\Delta E(t) = \Delta \mathcal{W}|_{\text{d}}(t) = \hbar\omega|\alpha_0|^2(e^{-2\kappa t} - 1) \leq 0$.

As another example, consider again a single cavity mode, this time prepared in its vacuum state $\rho_0 = |0\rangle\langle 0|$, that interacts with an outside bath in a squeezed-vacuum state [56] (Appendix C), eventually converging to a squeezed-vacuum state inside the cavity (Fig. 2b). Although the initial and the steady state have zero entropy, this is not true during the evolution (Fig. 3). Consequently, both dissipative passive-energy change $\Delta E_{\text{pas}}|_{\text{d}}(t)$ and dissipative ergotropy change $\Delta \mathcal{W}|_{\text{d}}(t) \geq 0$ occur. Figuratively, this process corresponds to a non-unitary charging of a battery.

III. REVERSIBILITY CRITERION

In non-equilibrium thermodynamics, the accepted criterion for the irreversibility or reversibility of the system relaxation to its steady state is the non-negativity of the entropy production [3, 57]. For quantum systems in contact with (thermal or non-thermal) Markovian baths, Spohn [43] put forward an expression for the entropy production $\Sigma(t)$. Here, we are interested in relaxation to steady state, for which we define $\Sigma := \Sigma(\infty)$, satisfying (Appendix D)

$$\Sigma \geq 0, \quad (5)$$

where the equality sign is the reversibility condition. For a constant Hamiltonian, it evaluates to $\Sigma = S(\rho_0||\rho_{\text{ss}}) \geq 0$, where $S(\rho_0||\rho_{\text{ss}}) := k_{\text{B}} \text{Tr}[\rho_0(\ln \rho_0 - \ln \rho_{\text{ss}})]$ is the entropy of the system initialised in a state ρ_0 at $t = 0$ relative to the steady state ρ_{ss} to which it relaxes. For a slowly

time-varying Hamiltonian [8, 58], Eq. (5) gives rise to an inequality for the change $\Delta\mathcal{S}$ of the system (von Neumann) entropy, given in Appendix D.

The common [8, 17, 20–23, 30, 33–42] identification of Eq. (5) with the second law appears plausible for systems in contact with thermal baths: It then evaluates to $\Sigma = \Delta\mathcal{S} - \mathcal{E}_d/T \geq 0$, where \mathcal{E}_d is the dissipative change in the system energy defined in Eq. (2a) (in the limit $t \rightarrow \infty$).

Here we contend that although inequality (5) is a formally correct statement of the second law (under standard thermodynamic assumptions), it may not provide a meaningful estimate of $\Delta\mathcal{S}$ if a system is initialised in a non-passive state and/or interacts with a non-thermal bath. Physically, this is because, as discussed above, the exchanged energy \mathcal{E}_d may be non-zero even if the entropy does not change.

IV. ENTROPY CHANGE IN RELAXATION PROCESSES INVOLVING ERGOTROPY

Consider the decay of an initially non-passive state ρ_0 to a (passive) thermal state ρ_{th} via contact with a thermal bath at temperature T . Based on the decomposition (3), the reversibility condition (5) evaluates to (at $t \rightarrow \infty$)

$$\Delta\mathcal{S} \geq \frac{\mathcal{E}_d}{T} = \frac{\Delta E_{\text{pas}}|_d + \Delta\mathcal{W}|_d}{T}, \quad (6)$$

where both dissipative change in passive energy (4a) and dissipated ergotropy (4b) appear. In what follows we shall revise this inequality, which may greatly overestimate the actual entropy change. As shown below, a tight inequality for $\Delta\mathcal{S}$ is indispensable for correctly assessing the maximum efficiency of an engine.

A. Constant Hamiltonian

As we have seen, dissipative ergotropy change is not necessarily linked to a change in entropy. Therefore, the lower bound on $\Delta\mathcal{S}$ in Eq. (6) may be not tight (maximal). It is obtained from Spohn's inequality (5) for the relaxation of an initially non-passive state in a thermal bath. However, one may resort to the fact that the entropy \mathcal{S} is a state variable, so that $\Delta\mathcal{S} = \mathcal{S}(\rho_{\text{th}}) - \mathcal{S}(\rho_0)$ is path-independent, i.e., its value only depends on the initial state ρ_0 and the (passive) thermal steady state ρ_{th} . Hence, Spohn's inequality (5) may well be applied to alternative evolution paths from ρ_0 to ρ_{th} , giving rise to different inequalities for the *same* $\Delta\mathcal{S}$.

In particular, we now consider a path that does not involve any dissipation of ergotropy to the bath: Namely, one may start the process by performing a unitary transformation to the passive state, $\rho_0 \mapsto \pi_0$. Thereafter, this state is brought in contact with the thermal bath, yielding the steady-state solution ρ_{th} . Inequality (5) applied to

this alternative path yields

$$\Delta\mathcal{S} \geq \frac{\Delta E_{\text{pas}}|_d}{T}, \quad (7)$$

where $\Delta E_{\text{pas}}|_d$ is the same as in Eq. (6).

The steady state attained via contact with a thermal bath is passive, hence the system ergotropy must decrease as a result of the relaxation, $\Delta\mathcal{W}|_d = -\mathcal{W}_0 \leq 0$, where $\mathcal{W}_0 \geq 0$ is the initial ergotropy stored in the state ρ_0 . Hence, inequality (7) always entails inequality (6) and is thus a tighter and more relevant estimate of $\Delta\mathcal{S}$. This has a crucial consequence: If the initial state is non-passive, inequality (7) rules out the equality sign in inequality (6), so that the considered decay via contact with a thermal bath can never be reversible according to criterion (5).

We now consider the more general situation wherein the system is governed by a constant Hamiltonian and interacts with an arbitrary bath (that may not be parameterised by a temperature) until it reaches the steady state ρ_{ss} . In order to obtain an optimal (the tightest) inequality for the entropy change $\Delta\mathcal{S}$, we here instead of inequality (5) (Σ for such a bath is given in Appendix E) propose to adopt the mathematical relation

$$S(\pi_0||\pi_{\text{ss}}) \geq 0. \quad (8)$$

As shown in Appendix F, Eq. (8) provides generally a tight inequality for $\Delta\mathcal{S}$. The motivation for Eq. (8) is, as before, that the entropy of any state ρ is the same as that of its passive counterpart π . If π_{ss} is a thermal state, we recover Eq. (7).

B. Time-dependent Hamiltonian

We now allow the Hamiltonian $H(t)$ to slowly vary during the evolution [8]. Contrary to the case of a constant Hamiltonian, the dissipative passive-energy change (4a) and the ergotropy change (4b) in the r.h.s. of Eq. (6) are now path-dependent. Namely, they are not only determined by the initial state ρ_0 and the steady state $\rho_{\text{th}}(\infty)$, which is a thermal state under the Hamiltonian $H(\infty)$.

Since during the evolution the time-dependent Hamiltonian may generate a non-passive state (even if the initial state is passive and the bath is thermal) we cannot, in general, find an alternative path void of dissipated ergotropy for the same $H(t)$. Notwithstanding, we may still consider a path void of *initial* ergotropy in the spirit of the previous section by extracting the ergotropy of the initial state in a *unitary* fashion prior to the interaction with the bath, resulting in the passive state π_0 . Afterwards, this passive state is brought into contact with the thermal bath, yielding the steady state $\rho_{\text{th}}(\infty)$. Spohn's inequality can be applied to the latter step, yielding

$$\Delta\mathcal{S} \geq \frac{\mathcal{E}'_d}{T}, \quad (9)$$

with the energy

$$\mathcal{E}'_d := \int_0^\infty \text{Tr}[\dot{\varrho}(t)H(t)]dt \quad (10)$$

exchanged with the bath along the alternative path. Here $\varrho(t)$ is the solution of the same thermal master equation that governs $\rho(t)$ but with the initial condition $\varrho_0 = \pi_0$. In the case that the initial state ρ_0 is already passive, we have $\varrho(t) = \rho(t)$, $\mathcal{E}'_d = \mathcal{E}_d$ and Eqs. (9) and (5) coincide. For a constant Hamiltonian, Eq. (9) evaluates to Eq. (7).

Consider now the more general situation where a quantum system interacts with a non-thermal bath and eventually relaxes to a unitarily-transformed thermal state $U\rho_{\text{th}}(\infty)U^\dagger$. A prime example is a harmonic oscillator that interacts with a squeezed thermal bath [56, 59]: Its steady state is a squeezed thermal state. Then one can show (Appendix G) that this situation can be traced back to the interaction of a unitarily-transformed state $\tilde{\rho}(t) := U^\dagger\rho(t)U$ with a *thermal* bath, provided that the Hamiltonian $H(t)$ commutes with itself at all times; a harmonic oscillator with a time-dependent frequency and time-independent eigenstates is an example. This requirement will be adopted in the remainder of this paper for any interaction of a system with a non-thermal bath. The relaxation of a possibly non-passive state $\tilde{\rho}(t)$ in a thermal bath pertains to the scenario considered above upon replacing $\rho(t)$ by $\tilde{\rho}(t)$ there. Equation (9) thus also holds for this class of non-thermal baths (the generalisation to arbitrary non-thermal baths is discussed in Appendix H).

The new entropic inequality (9) is the second main result of our work. For the special case of a constant Hamiltonian, it reduces to inequality (7).

V. MAXIMAL EFFICIENCY OF ENGINES POWERED BY NON-THERMAL BATHS

In view of our new inequality (9), does inequality (5) always provide a true bound on the engine efficiency? Namely, is reversibility indeed the key to operating a quantum engine at the highest possible efficiency? This question arises for cyclic engines fuelled by non-thermal (e.g., squeezed) baths, since such baths may transfer both passive thermal energy and ergotropy to the system while Eq. (5) does not distinguish between these two different kinds of energies.

Here we consider a quantum engine (Fig. 4) that operates between a cold thermal bath (at temperature T_c) and a hot non-thermal bath (whose thermal component has the temperature parameter $T_h > T_c$) subject to a time-dependent drive (the “piston” [8]). Existing treatments of such engines have taken the interactions with the baths to be isochoric, i.e., subject to a constant system Hamiltonian [25–30]. We here relax this restriction and allow for stroke cycles wherein the working-medium (WM) Hamiltonian is allowed to slowly change during the interaction with the baths [8]. More restrictively, during the interaction with the non-thermal bath, the Hamiltonian is

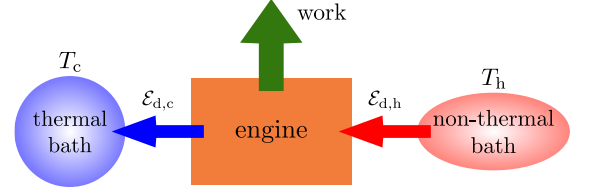


Figure 4. Schematics of an engine fuelled by a hot non-thermal (e.g., squeezed thermal) bath that provides the input energy $\mathcal{E}_{d,h}$. The engine operates in an arbitrary cycle wherein work is extracted by a piston and an amount of energy $\mathcal{E}_{d,c}$ is dumped into the cold thermal bath.

assumed to commute with itself at all times, as previously mentioned in the discussion on the validity of Eq. (9) for such a bath. We only impose the condition that the WM attains its steady state at the end of the energising stroke (wherein it interacts with the hot non-thermal bath) and the resetting stroke (wherein it interacts with the cold thermal bath).

The energising stroke is described by a master equation [42] that evolves the WM state to a unitarily-transformed thermal state $\rho_{ss}(\infty) = U\rho_{\text{th}}(\infty)U^\dagger$, hence Eq. (9) holds. After this stroke, the WM is in a non-passive state, whose ergotropy is subsequently extracted by the piston via a suitable unitary transformation. Since we seek the efficiency bound, we assume that no ergotropy is dissipated in the cold bath (and thus lost), hence the requirement to extract it from the WM before its interaction with that bath. We note that in cycles where both baths are simultaneously coupled to the WM (as in continuous cycles [60]), part of the ergotropy is inevitably dissipated into the cold bath, so that such cycles are inherently less efficient than stroke cycles adhering to the above requirement.

The engine’s WM must return to its initial state after each cycle. This implies that $\Delta S = 0$ over a cycle, hence the importance of having a tight estimate for the entropy change within each stroke. The entropy changes in the two relevant strokes satisfy $\Delta S_c \geq \mathcal{E}_{d,c}/T_c$ and $\Delta S_h \geq \mathcal{E}'_{d,h}/T_h$. Here $\mathcal{E}_{d,c} \leq 0$ is the change in the WM energy due to its interaction with the cold thermal bath and $\mathcal{E}'_{d,h} \geq 0$ is the change the WM energy would have, had the non-thermal bath been thermal [as in Eq. (9)]. Taking into account that the WM is passive prior to its interaction with the cold bath, so that Eqs. (9) and (5) coincide for that stroke, the condition of vanishing entropy change over a cycle (which must hold in any cycle) then yields the inequality

$$\Delta S_c + \Delta S_h = 0 \quad \Rightarrow \quad \frac{\mathcal{E}_{d,c}}{T_c} + \frac{\mathcal{E}'_{d,h}}{T_h} \leq 0. \quad (11)$$

The efficiency of the engine is defined as the ratio of the extracted work to the invested energy, $\eta := -W/\mathcal{E}_{d,h}$, where $\mathcal{E}_{d,h}$ is the total energy (the sum of passive thermal energy and ergotropy) imparted by the non-thermal bath during the energising stroke. Using the first-law statement (1), this ratio may be expressed through the energy

transfers $\mathcal{E}_{d,c}$ and $\mathcal{E}'_{d,h}$ (see Appendix I). Condition (11) on these energy transfers then restricts the efficiency to

$$\eta \leq 1 - \frac{T_c}{T_h} \frac{\mathcal{E}'_{d,h}}{\mathcal{E}_{d,h}} =: \eta_{\max}. \quad (12)$$

The bound (12) underscores the physicality of our inequality (9): In the usual regime of functioning of the engine, $\mathcal{E}'_{d,h} \geq 0$ and $\mathcal{E}_{d,h} > 0$ (i.e., the hot bath provides energy and increases the WM entropy), the bound (12) is limited by unity, $\eta_{\max} \leq 1$, which is reached in the “mechanical”-engine limit $\mathcal{E}'_{d,h} \rightarrow 0$ where the non-thermal bath only provides ergotropy. By contrast, the bound η_{Σ} that stems from the reversibility condition (5) (derived in Appendix I) may surpass 1 (see Ref. [30]). In the opposite, heat-engine, limit $\mathcal{E}'_{d,h} \rightarrow \mathcal{E}_{d,h}$ where only passive thermal energy but no ergotropy is imparted by the hot bath, Eq. (12) reproduces the Carnot bound $\eta_C = 1 - T_c/T_h$. As shown below, if the Hamiltonian is kept constant during the interaction with the non-thermal bath, then Eq. (12) is restricted by $\eta_C \leq \eta_{\max} \leq \eta_{\Sigma}$. Therefore, for such engines our new bound (12) is always tighter than the second-law bound η_{Σ} .

The bound (12) is valid in the regime $\mathcal{E}_{d,c} \leq 0$ and $\mathcal{E}'_{d,h} \geq 0$ wherein the cold bath serves as an energy dump. As shown in [29], there exists a regime wherein such a machine acts simultaneously as an engine and a refrigerator for the cold bath. The efficiency then evaluates to $\eta = 1$ (see Appendix I).

We have thus reached a central conclusion: The efficiency bound of the engine increases with the decrease of the ratio of the energy that an alternative thermal engine would have received (in the same energising stroke) to the total energy imparted by the non-thermal bath (in the actual engine cycle). In the limit of thermal baths [8] we recover the standard Carnot bound for the efficiency of heat engines, even if the engine (in any cycle) exhibits quantum signatures (e.g., quantum coherence in the WM due to the piston action [17]) or the WM–bath interactions are time-dependent [61].

VI. SPECIFIC QUANTUM ENGINES

We now pose the question: Which bound is more relevant, η_{Σ} (whose explicit form is given in Appendix I) that stems from the reversibility condition (5), or η_{\max} given by Eq. (12)? Contrary to the Carnot bound, the efficiency bound (12) not only depends on the parameters of the baths but also on the energising stroke through the stroke’s initial condition and the Hamiltonian that determine the integrals $\mathcal{E}'_{d,h}$ and $\mathcal{E}_{d,h}$. Yet, the functional form (12) is independent of the choice of the non-thermal bath or the WM. Whether or not this bound is reached by an engine that implements this chosen energising stroke is then determined by condition (11).

In complete generality, the tighter of the alternative

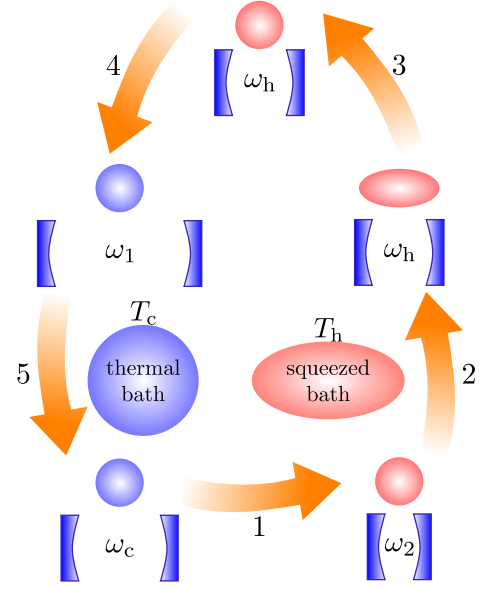


Figure 5. A photonic Carnot cycle for a squeezed thermal bath. The cycle starts with a thermal state with frequency ω_c and temperature T_c (lower left corner). In stroke 1, the mode undergoes an adiabatic compression to frequency $\omega_2 = \omega_c T_h / T_c$ and temperature $T_h > T_c$. Thereafter, in the energising stroke 2, the frequency is slowly reduced to $\omega_h \leq \omega_2$ while the mode is connected to the squeezed thermal bath, yielding a squeezed thermal steady state. Its ergotropy is extracted in stroke 3 by an “unsqueezing” unitary operation, resulting in a thermal state with temperature T_h . In stroke 4, the frequency is again adiabatically reduced to $\omega_1 = \omega_h T_c / T_h$ such that the mode attains the temperature T_c . Finally, stroke 5 is an isothermal compression back to the initial state.

efficiency bounds derived here,

$$\eta \leq \min\{\eta_{\max}, \eta_{\Sigma}\}, \quad (13)$$

is the relevant one. Relation (13) is the universal thermodynamic limit on quantum engine efficiency, which never surpasses unity.

Notwithstanding the alternatives that may be offered by Eq. (13), we now discuss two generic practically-relevant engine cycles for which one can explicitly show that $\eta_{\max} \leq \eta_{\Sigma}$. Such engines are thus not restricted by the second law, but by other constraints on their entropy.

A. Time-dependent Hamiltonian: A squeezed photonic Carnot engine

We consider a photonic Carnot-like engine fuelled by a squeezed-thermal bath, as depicted in Fig. 5. It contains the four strokes of the regular thermal Carnot cycle [1–4], as well as an additional ergotropy-extraction stroke (stroke 3 in the figure). In the regular thermal Carnot cycle, the interactions with the baths are isothermal.

Based on Eq. (9), we have in the second stroke $\mathcal{E}'_{d,h} = T_h \Delta \mathcal{S}_h$, since the master equation void of squeeze-

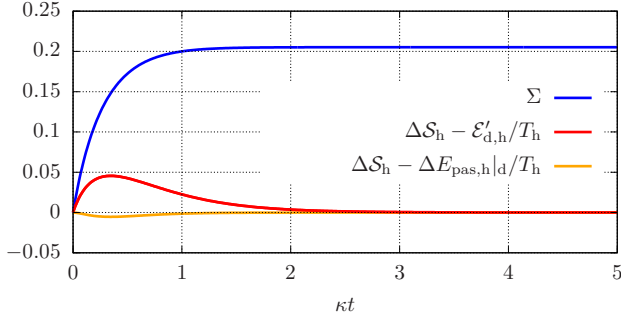


Figure 6. Change in entropy (in units of k_B) during stroke 2 of the modified Carnot cycle in Fig. 5 as a function of the stroke duration obtained by a numerical integration of the master equation. The upper (blue) curve corresponds to the reversibility criterion (5); it is seen that the inequality $\Sigma \geq 0$ is far from being saturated. By contrast, our proposed inequality (9) is saturated (i.e., the equality sign applies) for sufficiently long stroke duration (red central curve). Then, as seen from the lower (orange) curve, also $\mathcal{E}'_{d,h} = \Delta E_{pas,h}|_d$, due to the fact that the squeezing only occurs at the beginning of the stroke. Equation (9) here also holds for a finite cycle duration. Parameters: Oscillator frequency $\omega(t) = 25\kappa - 0.05\kappa t$, $k_B T_h = 5\hbar\kappa$ and squeezing parameter $r = 0.2$.

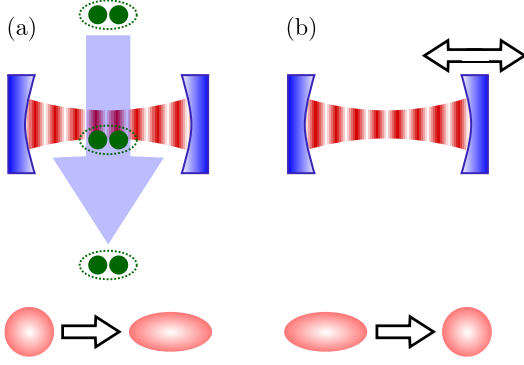


Figure 7. (a) The interaction of a cavity mode with a squeezed thermal bath (stroke 2 in Fig. 5) may be realised in a micro-maser setup where a beam of entangled atom pairs passes through the cavity [31]. (b) The unsqueezing operation in stroke 3 of Fig. 5 may be implemented by a suitable modulation of the cavity frequency [62–64].

ing induces isothermal expansion wherein the state $\varrho(t)$ is always in thermal equilibrium (Fig. 6). Stroke 5 is isothermal compression, i.e., $\mathcal{E}_{d,c} = T_c \Delta \mathcal{S}_c$. The condition of vanishing entropy change over a cycle, $\Delta \mathcal{S} = \mathcal{E}_{d,c}/T_c + \mathcal{E}'_{d,h}/T_h = 0$, corresponds to the equality sign in condition (11). Hence, the efficiency of this cycle is the bound (12).

Consequently, the bound η_{\max} is lower than η_Σ for all possible engine cycles that contain a “Carnot-like” energising stroke, namely, a stroke characterised by a slowly-changing Hamiltonian and an initial thermal state at temperature T_h , such that $\mathcal{E}'_{d,h} = T_h \Delta \mathcal{S}_h$.

Such a photonic Carnot engine energised by a squeezed bath may be implemented as a modification of the pho-

tonic Carnot cycle based on a cavity in a micromaser setup in the seminal work by Scully et al. [9]: Instead of a beam of coherently-prepared three-level atoms (“phaseonium”) that constitute an effective thermal bath for the cavity-mode WM, we here suggest, following Ref. [31], to use a beam of suitably-entangled atom pairs passing through a cavity that may act as a squeezed-thermal bath for the same WM (Fig. 7a). A major advantage of this method is that it allows for very high squeezing parameters. In order to extract the ergotropy that is stored in the cavity mode after its interaction with the squeezed bath and before its interaction with the cold bath (where it would be lost), a unitary transformation that “unsqueezes” the cavity field must be performed, e.g., as in Refs. [62–64], where the cavity-mode frequency is abruptly ramped up and then gradually ramped down (Fig. 7b).

B. Constant Hamiltonian: An Otto-like cycle

Next, we consider a quantum Otto cycle [17, 65–68] that consists of two isentropic strokes (adiabatic compression and decompression of the WM), two isochoric strokes (interaction with the baths at a fixed Hamiltonian) and an additional ergotropy-extraction stroke. This cycle amounts to setting $\omega_2 = \omega_h$ and $\omega_1 = \omega_c$ in Fig. 5.

Since the Hamiltonian is now kept constant during the energising stroke, we have $\mathcal{E}'_{d,h} = \Delta E_{pas,h}$, where $\Delta E_{pas,h}$ is the change in passive energy during the hot stroke, and $\mathcal{E}_{d,h} = \Delta E_{pas,h} + \Delta \mathcal{W}_h$, where $\Delta \mathcal{W}_h$ is the change in ergotropy during that stroke. The efficiency of this Otto-like cycle is bounded by Eq. (12),

$$\eta_{\max}^{\text{Otto}} = 1 - \frac{T_c}{T_h} \frac{\Delta E_{pas,h}}{\Delta E_{pas,h} + \Delta \mathcal{W}_h} \leq \eta_\Sigma, \quad (14)$$

but this bound is only attained in the “mechanical” limit $\mathcal{E}'_{d,h} = \Delta E_{pas,h} = 0$, where only ergotropy is transferred from the non-thermal bath and no net entropy change occurs during the strokes. By contrast, the Carnot-like cycle always operates at maximum efficiency, even when both passive thermal energy and ergotropy are imparted by this bath.

In general, any engine cycle wherein the interaction with the hot bath is isochoric (has constant Hamiltonian) and sufficiently long (for the WM to reach steady state) abides by the bound (14), which is lower than the bound η_Σ imposed by the second law (Fig. 8). Moreover, their efficiency bound always surpasses the Carnot bound, $\eta_{\max}^{\text{Otto}} \geq \eta_C$.

VII. CONCLUSIONS

Our analysis has been aimed at comparing the efficiency bounds and the conditions for their attainment in quantum engines energised by thermal and non-thermal baths. These respective bounds turn out to be very different

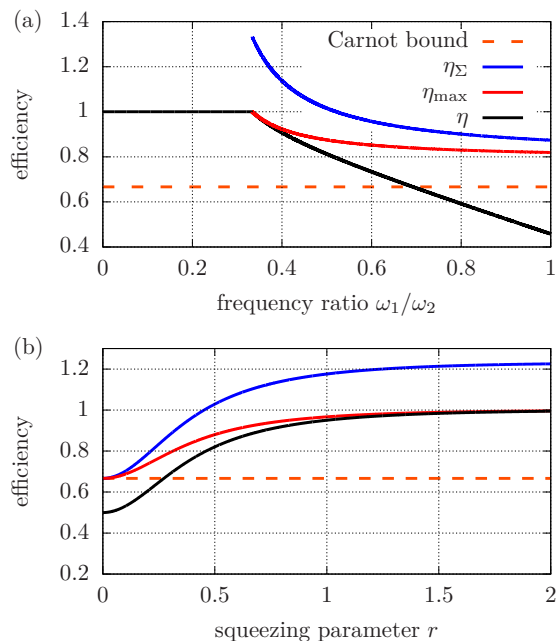


Figure 8. Actual efficiency η and alternative efficiency bounds (the explicit expressions are summarised in Appendix J) for an Otto-like cycle implemented with a harmonic-oscillator working medium and a squeezed thermal bath as a function of (a) the frequency ratio and (b) the squeezing parameter. The bounds only hold in the regime $\mathcal{E}_{d,c} \leq 0$ (see text). Parameters: $T_h = 3T_c$ and (a) squeezing parameter $r = 0.5$ and (b) oscillator frequencies $\omega_h = 2\omega_c$.

since, unlike thermal baths, non-thermal baths may exchange thermal (passive) energy and ergotropy with the working medium (WM). To this end we have revisited the first law of thermodynamics and identified as passive-energy change the part of the energy exchange with the bath that *necessarily* causes a change in the WM entropy [Eq. (4a)]. This division of the exchanged energy relies on the distinction between passive and non-passive states of the WM. Only the latter states store ergotropy that may be completely extracted in the form of work. We note that our approach based on non-passive states conceptually differs from the energetic division involving “housekeeping heat” previously provided for classical systems [69].

Based on the distinction between passive and non-passive states, we have put forward a new estimate (9) of the entropy change in quantum relaxation processes, which turns out to be the key for understanding the limitations of quantum engines fuelled by arbitrary baths. Cyclic engines whose passive energy is altered by the baths are restricted in efficiency by limits on their entropy change. Yet, for a wide class of practically-relevant engines, including all engines whose energising stroke is either isochoric or Carnot-like, the restriction imposed by inequality (9) on the entropy change is stricter than what the second law (5) would allow. By contrast, the commonly used reversibility is a global condition on the WM

and the baths combined that is imposed by the second law, and hence not necessarily a relevant characterisation of engine efficiency.

An alternative formulation of our main insight is that, for any baths, entropy change limits the engine efficiency in the *same* way as in traditional heat engines—condition (11) is the same whether the energising bath is thermal or not. Namely, maximal efficiency is reached when (a) no ergotropy (extractable work) is dumped into the cold bath and (b) no entropy is generated within the engine, or, equivalently, minimal energy is dumped into the cold bath [3]. For thermal engines, this criterion of minimal energy dumping and the reversibility criterion coincide, but the two criteria differ if the energising bath is non-thermal.

Another important insight is that the same efficiency bound (12) ensues whether the WM is energised by a non-thermal bath or by a thermal bath (that supplies thermal energy) combined with a battery (that supplies ergotropy) provided the total energy imparted by the WM remains the same. This supports the description of non-thermal engines as hybrids of thermal (thermal-energy-fuelled) and “mechanical” (ergotropy-fuelled) engines [29].

Our theory provides better understanding of the operation principles of quantum engines: These are shown not to follow only from the laws of thermodynamics, but require discrimination between different (passive and non-passive) quantum states of the system (WM) and the baths involved. The present generalisation of the treatment of standard thermal processes for quantum systems is not only the key to the construction of the most efficient hybrid engines that are unrestricted by the Carnot bound. It may also open a new perspective on quantum-channel communications [52, 55, 70] where entropic constraints play a key role.

ACKNOWLEDGEMENTS

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Appendix A: Non-passive states and maximally extractable work

The energy E of a state ρ with respect to a Hamiltonian H can be decomposed into ergotropy \mathcal{W} and passive energy E_{pas} . Ergotropy is the maximum amount of work that can be extracted from the state by means of cyclic unitary transformations, meaning that the Hamiltonian before and after the unitary coincide [6, 7, 10]. The passive energy, by contrast, cannot be extracted in the form of work. States that only contain passive energy are called *passive states*.

Ergotropy is defined as

$$\mathcal{W}(\rho, H) := \text{Tr}(\rho H) - \min_U \text{Tr}(U \rho U^\dagger H) \geq 0, \quad (\text{A1})$$

where the minimisation is over the set of all possible unitary transformations. Consequently, any state ρ can be written as $\rho = V_\rho \pi V_\rho^\dagger$, i.e., as a unitarily-transformed passive state π , where V_ρ is the unitary that realises the minimum appearing on the r.h.s. of Eq. (A1). The energy of the state ρ thus reads

$$E = E_{\text{pas}} + \mathcal{W} = \text{Tr}[\pi H] + \text{Tr}[(\rho - \pi)H]. \quad (\text{A2})$$

Explicitly, the passive state and its energy read

$$\pi := \sum_n r_n |n\rangle\langle n| \quad (\text{A3a})$$

$$E_{\text{pas}} = \text{Tr}[\pi H] = \sum_n r_n E_n, \quad (\text{A3b})$$

where $\{r_n\}$ are the ordered ($r_{n+1} \leq r_n \forall n$) eigenvalues of ρ and $\{|n\rangle\}$ is the ordered ($E_{n+1} \geq E_n \forall n$) eigenbasis of H . When H is non-degenerate, π is unique. If H is degenerate, its eigenbasis and, consequently, the passive state (A3a), may be not unique. However, the energies (A3b) of all passive states corresponding to ρ are the same and equal the passive energy of ρ .

Appendix B: Signs of $\Delta E_{\text{pas}|d}$ and $\Delta \mathcal{S}$ under a majorisation relation

Assume $\rho(t') \succ \rho(t'')$ for any $t'' \geq t'$ in some time interval I ($t', t'' \in I$), namely that $\rho(t')$ majorises [40, 55] $\rho(t'')$ in this interval, i.e.,

$$\sum_{m=1}^n r_m(t') \geq \sum_{m=1}^n r_m(t'') \quad (1 \leq n \leq N), \quad (\text{B1})$$

where $r_{m+1}(\tau) \leq r_m(\tau)$ ($\tau \in I$) are the ordered eigenvalues of $\rho(\tau)$ [cf. Eq. (A3a)] and N is the dimension of the Hilbert space of the system.

Let us consider the sign of the dissipative passive-energy change $\Delta E_{\text{pas}|d}$ under this majorisation condition. We may write (4a) in the form

$$\Delta E_{\text{pas}|d}(t) = \int_0^t d\tau \text{Tr}[\dot{\pi}(\tau)H(\tau)] = \int_0^t d\tau \lim_{h \rightarrow 0} f(\tau, h), \quad (\text{B2})$$

where we have defined

$$f(\tau, h) := \sum_{n=1}^N \frac{r_n(\tau + h) - r_n(\tau)}{h} E_n(\tau), \quad (\text{B3})$$

where $E_{n+1}(\tau) \geq E_n(\tau)$ are the ordered eigenvalues of the Hamiltonian [cf. Eq. (A3b)]. Using summation by parts and the normalisation of the density matrix, this function may be rewritten as

$$f(\tau, h) = \sum_{n=1}^{N-1} [E_{n+1}(\tau) - E_n(\tau)] \sum_{m=1}^n \frac{r_m(\tau) - r_m(\tau + h)}{h}. \quad (\text{B4})$$

The first factor is non-negative due to the monotonically-ordered energies. The second factor is also non-negative if Eq. (B1) holds in the entire integration domain $[0, t]$. In this case, the majorisation relation implies $\Delta E_{\text{pas}|d}(t) \geq 0$.

Let us now turn to the sign of the entropy change. If $\rho_1 \succ \rho_2$, then $\mathcal{S}(\rho_2) \geq \mathcal{S}(\rho_1)$ [10]. Hence, we have the relation

$$\begin{aligned} \rho(t') \succ \rho(t'') \quad \forall 0 \leq t' \leq t'' \leq t \\ \Rightarrow \quad \Delta E_{\text{pas}|d}(t) \geq 0 \wedge \Delta \mathcal{S}(t) \geq 0, \end{aligned} \quad (\text{B5})$$

where $\Delta \mathcal{S}(t) = \mathcal{S}(\rho(t)) - \mathcal{S}(\rho_0)$. Similarly, one can show that the opposite relation holds, $\rho(t') \prec \rho(t'') \Rightarrow \Delta E_{\text{pas}|d}(t) \leq 0 \wedge \Delta \mathcal{S}(t) \leq 0$. When the Hamiltonian is non-degenerate, $\Delta E_{\text{pas}|d}(t)$ and $\Delta \mathcal{S}(t)$ can be shown to vanish iff the passive state corresponding to $\rho(\tau)$ is constant (i.e., the evolution of $\rho(\tau)$ is unitary) for $\tau \in [0, t]$.

For the case of a constant Hamiltonian, relation (B5) was obtained in Ref. [40]. In this case, $\Delta E_{\text{pas}|d}(t) = \Delta E_{\text{pas}}(t)$ and hence Eq. (B5) implies that the passive energy of ρ_2 is greater than or equal to the passive energy of ρ_1 if $\rho_1 \succ \rho_2$ or, equivalently, if $\pi_1 \succ \pi_2$, where π_i is the passive state corresponding to ρ_i ($i = 1, 2$).

Appendix C: Master equation for a squeezed bath

In the interaction picture, the master equation for a harmonic oscillator that interacts with a squeezed thermal bath reads [56]

$$\begin{aligned} \dot{\rho} = \kappa(N+1)\mathcal{D}(a, a^\dagger)[\rho] + \kappa N\mathcal{D}(a^\dagger, a)[\rho] \\ - \kappa M\mathcal{D}(a, a)[\rho] - \kappa M\mathcal{D}(a^\dagger, a^\dagger)[\rho], \end{aligned} \quad (\text{C1})$$

where $\mathcal{D}(A, B)[\rho] := 2A\rho B - B A \rho - \rho B A$. Here κ denotes the decay rate and (w.l.o.g. we have set the squeezing phase to zero)

$$N := \bar{n}(\cosh^2 r + \sinh^2 r) + \sinh^2 r \quad (\text{C2a})$$

$$M := -\cosh r \sinh r (2\bar{n} + 1), \quad (\text{C2b})$$

where $\bar{n} = [\exp(\hbar\omega/[k_B T]) - 1]^{-1}$ is the thermal excitation number of the bath at the oscillator frequency ω and r the squeezing parameter. The results in Fig. 3 were obtained by a numerical solution of Eq. (C1) with $\bar{n} = 0$.

Defining $b := S(r)aS^\dagger(r) = a \cosh r + a^\dagger \sinh r$, where $S(r) = \exp[\frac{r}{2}a^2 - \frac{r}{2}(a^\dagger)^2]$ is the unitary squeezing operator, the master equation (C1) can be cast into the Lindblad form [42, 59]

$$\dot{\rho} = \kappa(\bar{n} + 1)\mathcal{D}(b, b^\dagger)[\rho] + \kappa\bar{n}\mathcal{D}(b^\dagger, b)[\rho]. \quad (\text{C3})$$

Its steady-state solution is the squeezed thermal state $S(r)[Z^{-1} \exp(-\hbar\omega a^\dagger a/[k_B T])]S^\dagger(r)$.

Appendix D: Entropy production Σ

Spohn's inequality for the entropy-production rate reads [43]

$$\sigma := -\frac{d}{dt} S(\rho(t) \parallel \rho_{ss}) \geq 0, \quad (\text{D1})$$

where $S(\rho(t) \parallel \rho_{ss}) := k_B \text{Tr}[\rho(t)(\ln \rho(t) - \ln \rho_{ss})]$. Inequality (D1) holds for any $\rho(t)$ that evolves according to a Lindblad master equation [42]

$$\dot{\rho} = \mathcal{L}\rho, \quad (\text{D2})$$

\mathcal{L} being the Liouvillian (Lindblad operator). Its steady-state solution obeys $\mathcal{L}\rho_{ss} = 0$. Then, upon defining $\Sigma := \int_0^\infty \sigma dt$, the time-integrated inequality (D1) yields

$$\Sigma = S(\rho_0 \parallel \rho_{ss}) \geq 0. \quad (\text{D3})$$

Note that Eq. (D1) may not hold for non-Markovian baths [11].

As shown in Refs. [8, 58], Spohn's inequality (D1) can be generalised to time-dependent Hamiltonians under the condition that $H(t)$ varies slowly compared to the relaxation time of the reservoir [8]. The corresponding master equation then reads

$$\dot{\rho}(t) = \mathcal{L}(t)\rho(t), \quad (\text{D4})$$

where $\mathcal{L}(t)$ is the same Liouvillian as in Eq. (D2), but with time-dependent coefficients (cf. Ref. [8]). Its invariant state $\rho_{ss}(t)$ satisfies $\mathcal{L}(t)\rho_{ss}(t) = 0$. The generalisation of inequality (D1) then reads [58]

$$\sigma = -\frac{d}{ds} S\left(e^{s\mathcal{L}(t)}\rho(t) \parallel \rho_{ss}(t)\right) \Big|_{s=0} \geq 0. \quad (\text{D5})$$

Upon integration, Eq. (D5) evaluates to the inequality

$$\Sigma = \Delta S + k_B \int_0^\infty \text{Tr} \left[(\mathcal{L}(t)\rho(t)) \ln \rho_{ss}(t) \right] dt \geq 0 \quad (\text{D6})$$

for the entropy change $\Delta S = S(\rho_{ss}(\infty)) - S(\rho_0)$. In the case of a constant Hamiltonian, Eq. (D6) reduces to Eq. (D3).

If the Liouvillian describes the interaction with a thermal bath at temperature T , i.e., $\mathcal{L}(t) = \mathcal{L}_{th}(t)$, then $\rho_{ss}(t) = \rho_{th}(t)$, where

$$\rho_{th}(t) = \frac{1}{Z(t)} \exp\left(-\frac{H(t)}{k_B T}\right) \quad (\text{D7})$$

is a thermal state for the (instantaneous) Hamiltonian $H(t)$. Equation (D6) then yields

$$\Delta S \geq \frac{1}{T} \int_0^\infty \text{Tr} [\dot{\rho}(t)H(t)] dt = \frac{\mathcal{E}_d}{T}, \quad (\text{D8})$$

with the dissipated energy \mathcal{E}_d defined in Eq. (2a).

Appendix E: Entropy production Σ for non-thermal baths

Let us consider Σ in the case of a constant Hamiltonian [Eq. (D3)] for a non-thermal bath that gives rise to a non-passive steady state $\rho_{ss} = U\pi_{ss}U^\dagger$ via the Liouvillian \mathcal{L}_U . This Σ can be related to that of a passive state, as follows. Since the relative entropy is invariant with respect to a unitary transformation of its arguments, Eq. (D3) can be recast in the form

$$\Sigma = S(\tilde{\rho}_0 \parallel \pi_{ss}) \geq 0, \quad (\text{E1})$$

where $\tilde{\rho}_0 := U^\dagger \rho_0 U$. Thus, Σ equals the entropy production obtained under the relaxation of an open system from the unitarily-transformed state $\tilde{\rho}_0$ to the passive state π_{ss} .

In particular, when π_{ss} is the thermal state ρ_{th} , Σ equals the entropy production obtained under thermalisation of the system starting from the state $\tilde{\rho}_0$ and we have

$$\Sigma = \Delta S - \frac{\tilde{\mathcal{E}}_d}{T} \geq 0, \quad (\text{E2})$$

where $\tilde{\mathcal{E}}_d$ is the change in the energy $\tilde{E} = \text{Tr}[\tilde{\rho}H]$ of the transformed state $\tilde{\rho}$.

Consider now a slowly-varying $H(t)$ such that inequality (D6) holds. The invariant state of $\mathcal{L}_U(t)$ now reads $\rho_{ss}(t) = U\rho_{th}(t)U^\dagger$, with the (instantaneous) thermal state (D7). Inequality (D6) then yields

$$\Sigma = \Delta S - \frac{1}{T} \int_0^\infty \text{Tr} [U^\dagger \dot{\rho}(t)U H(t)] dt \geq 0, \quad (\text{E3})$$

where the appearing integral is the generalisation of $\tilde{\mathcal{E}}_d$ from inequality (E2). It is shown in Appendix G that $U^\dagger \dot{\rho}(t)U$ equals a thermal Liouvillian acting on a unitarily-transformed state [Eq. (G6)]. Hence, also for a time-dependent Hamiltonian, the evaluation of Σ in a non-thermal bath reduces to the case of a transformed state that decays via contact with a thermal bath.

Appendix F: Optimality of inequality (8)

Equation (8) provides a generally tighter inequality for ΔS than Eq. (E1) [or (D3)]. Indeed, Eq. (E1) can be written as $\Delta S \geq S(\pi_{ss}) - k_B A$, where $A = -\text{Tr}[\tilde{\rho}_0 \ln \pi_{ss}]$. This inequality is the tightest (i.e., its r.h.s. is maximal) on the set of all states $\tilde{\rho}_0$ which differ from ρ_0 by a unitary transformation, when A is minimal on this set. Note that π_{ss} commutes with the Hamiltonian and the eigenvalues of $-\ln \pi_{ss}$ do not decrease as a function of the eigenvalues of the Hamiltonian. Thus, $-\ln \pi_{ss}$ can be considered, in a sense, as an effective ‘‘Hamiltonian’’, for which A is the average ‘‘energy’’ in the state $\tilde{\rho}_0$. The average energy amongst unitarily-accessible states is known to be minimised in the passive state. When H is non-degenerate, then the passive state π_0 corresponding to H is also the

passive state corresponding to the effective “Hamiltonian” $-\ln \pi_{ss}$; hence, A is minimal for $\tilde{\rho}_0 = \pi_0$.

By contrast, if H is degenerate there is generally no unique passive state (Appendix A). In this case, A is minimal not for each π_0 but iff π_0 is also a passive state of the effective “Hamiltonian”, i.e., iff π_0 commutes with π_{ss} . One can show that there exists, at least, one such state π_0 . Thus, Eq. (8) provides the tightest inequality for ΔS among all inequalities of the form (E1) or (D3).

Appendix G: Unitary equivalence of non-thermal and thermal baths

The time evolution of an initial state ρ_0 under the Liouvillian \mathcal{L}_U as defined in Appendix E may be replaced by an alternative time evolution involving a *thermal* bath. These two equivalent evolution paths can be lucidly represented by the diagram (see also [59] and Appendix C)

$$\begin{array}{ccc} \rho_0 & \xrightarrow{\mathcal{L}_U} & \rho(t) = U\tilde{\rho}(t)U^\dagger \\ \uparrow U^\dagger & & \downarrow U \\ \tilde{\rho}_0 = U^\dagger \rho_0 U & \xrightarrow{\mathcal{L}_{th}} & \tilde{\rho}(t) \end{array} \quad (G1)$$

According to Eq. (G1), the evolution of ρ_0 induced by a non-thermal bath towards ρ_{ss} (solid arrow) may be replaced by a three-stage process (dashed arrows) wherein the system is in contact with a thermal bath only in the second step.

This may be shown as follows. The Liouvillian \mathcal{L}_U in the interaction picture may be cast into the general Lindblad form [42]

$$\mathcal{L}_U \rho = \sum_{\alpha} \frac{\gamma_{\alpha}}{2} [2L_{\alpha} \rho L_{\alpha}^\dagger - L_{\alpha}^\dagger L_{\alpha} \rho - \rho L_{\alpha}^\dagger L_{\alpha}]. \quad (G2)$$

We now consider the unitarily transformed master equation

$$U^\dagger (\mathcal{L}_U \rho) U = \sum_{\alpha} \frac{\gamma_{\alpha}}{2} [2\tilde{L}_{\alpha} \tilde{\rho} \tilde{L}_{\alpha}^\dagger - \tilde{L}_{\alpha}^\dagger \tilde{L}_{\alpha} \tilde{\rho} - \tilde{\rho} \tilde{L}_{\alpha}^\dagger \tilde{L}_{\alpha}], \quad (G3)$$

where we have defined $\tilde{\rho} := U^\dagger \rho U$ and $\tilde{L}_{\alpha} := U^\dagger L_{\alpha} U$. The right-hand side of Eq. (G3) is thus again a Lindblad superoperator, $U^\dagger (\mathcal{L}_U \rho) U =: \tilde{\mathcal{L}} \tilde{\rho}$. Now, since $\rho_{ss} = U \rho_{th} U^\dagger$ is the steady-state solution of \mathcal{L}_U , the state $\tilde{\rho}_{ss} := U^\dagger \rho_{ss} U = \rho_{th}$ must be the steady state of $\tilde{\mathcal{L}}$. Hence, $\tilde{\mathcal{L}}$ has to be a thermal generator, i.e., $\tilde{\mathcal{L}} = \mathcal{L}_{th}$, and therefore

$$U^\dagger (\mathcal{L}_U \rho) U = \mathcal{L}_{th} (U^\dagger \rho U). \quad (G4)$$

Hence, the solution of $\dot{\rho} = \mathcal{L}_U \rho$ may be written as

$$\rho(t) = U [e^{t\mathcal{L}_{th}} (U^\dagger \rho_0 U)] U^\dagger. \quad (G5)$$

If $H(t)$ is slowly varying in time and commutes with itself at all times, we have time-dependent $\gamma_{\alpha}(t)$ in

Eq. (G2) [8]. Since the above derivation does not depend on these rates, we have

$$U^\dagger (\mathcal{L}_U(t) \rho(t)) U = \mathcal{L}_{th}(t) (U^\dagger \rho(t) U). \quad (G6)$$

Appendix H: Validity of inequality (9) for non-thermal baths

Equation (9) for a thermal bath was derived based on the alternative (dashed) path

$$\begin{array}{ccc} \rho_0 & \xrightarrow[\mathcal{E}_d]{\dot{\rho}(t)=\mathcal{L}_{th}(t)\rho(t)} & \rho_{th}(\infty) \\ \uparrow \text{unitary} & & \downarrow \mathcal{E}'_d \\ \rho_0 = \pi_0 & \xrightarrow[\mathcal{E}'_d]{\dot{\varrho}(t)=\mathcal{L}_{th}(t)\varrho(t)} & \varrho(t) \end{array} \quad (H1)$$

The energies \mathcal{E}_d (along the original path) and \mathcal{E}'_d (along the alternative path) are those that appear on the r.h.s. of the entropic inequalities (6) and (9).

The Σ -inequality for the situation where the invariant state is non-passive is given in Eq. (E3) and may be recast in the form

$$\Delta S \geq \frac{1}{T} \int_0^\infty \text{Tr} [U^\dagger [\mathcal{L}_U(t) \rho(t)] U H(t)] dt. \quad (H2)$$

Owing to Eq. (G6), this inequality is equivalent to

$$\Delta S \geq \frac{1}{T} \int_0^\infty \text{Tr} [\mathcal{L}_{th}(t) \tilde{\rho}(t)] H(t) dt, \quad (H3)$$

where $\tilde{\rho}(t) := U^\dagger \rho(t) U$ and $\mathcal{L}_{th}(t)$ is a thermal Liouvillian with the same temperature and the same $H(t)$ as in $\mathcal{L}_U(t)$. The problem of a state $\rho(t)$ that evolves subject to a non-thermal bath has thus been reduced to the problem of a state $\tilde{\rho}(t)$ that evolves according to a thermal bath. This is the situation considered in the original (solid) path in Eq. (H1) upon replacing $\rho(t)$ by $\tilde{\rho}(t)$ there. This yields Eq. (9), thus extending it to the case of a non-passive invariant state.

In the general case that $\pi_{ss}(t)$ is not a thermal state, inequality (H2) is replaced by

$$\Delta S \geq -k_B \int_0^\infty \text{Tr} [U^\dagger [\mathcal{L}_U(t) \rho(t)] U \ln \pi_{ss}(t)] dt. \quad (H4)$$

One can then proceed as above, but $\mathcal{L}_{th}(t)$ is then replaced by a “passive” Liouvillian $\mathcal{L}_{pas}(t)$ whose invariant state is $\pi_{ss}(t)$. The resulting inequality for ΔS [the generalisation of Eq. (9), i.e., the counterpart of Eq. (8)] then reads,

$$\Delta S \geq -k_B \int_0^\infty \text{Tr} [\mathcal{L}_{pas}(t) \varrho(t)] \ln \pi_{ss}(t) dt, \quad (H5)$$

where $\varrho(0) = \pi_0$. Note that the latter integral cannot be identified with energy transfer. Equation (H5) holds also for the case of a passive invariant state $\rho_{ss}(t) = \pi_{ss}(t)$, where now $\mathcal{L}_{pas}(t) = \mathcal{L}(t)$.

Appendix I: Derivation of the efficiency bound

Energy conservation [Eq. (1)] over a cycle yields

$$\mathcal{E}_{d,c} + \mathcal{E}_{d,h} + W = 0, \quad (\text{I1})$$

where $\mathcal{E}_{d,c}$ ($\mathcal{E}_{d,h}$) is the dissipative energy change of the WM due to its interaction with the cold thermal (hot non-thermal) bath. As mentioned in the main text, we assume that the WM is thermal and hence passive prior to its interaction with the cold thermal bath.

The efficiency of the engine is defined as the ratio of the extracted work to the invested energy (passive thermal energy and ergotropy) $\mathcal{E}_{d,h} = \int_0^\infty \text{Tr}[(\mathcal{L}_U(t)\rho(t))H(t)]dt$ provided by the non-thermal bath, yielding

$$\eta = \frac{-W}{\mathcal{E}_{d,h}} = 1 + \frac{\mathcal{E}_{d,c}}{\mathcal{E}_{d,h}}. \quad (\text{I2})$$

This expression holds for $\mathcal{E}_{d,c} \leq 0$ and $\mathcal{E}_{d,h} \geq 0$; see below a discussion of the opposite case.

According to the reversibility condition (5), the requirement of vanishing entropy change over a cycle then yields

$$\frac{\mathcal{E}_{d,c}}{T_c} + \frac{\tilde{\mathcal{E}}_{d,h}}{T_h} \leq 0, \quad (\text{I3})$$

where $\tilde{\mathcal{E}}_{d,h}$ [the integral in Eq. (E3)] is the energy change during the interaction with the thermal bath along the dashed path in Eq. (G1). Consequently, according to this criterion the efficiency is bounded by

$$\eta \leq 1 - \frac{T_c}{T_h} \frac{\tilde{\mathcal{E}}_{d,h}}{\mathcal{E}_{d,h}} =: \eta_\Sigma. \quad (\text{I4})$$

This bound surpasses 1 if $\tilde{\mathcal{E}}_{d,h} < 0$, which, e.g., is the case if the bath is “over-squeezed”: This means that, due to the excessive bath squeezing, the interaction with the thermal

bath along the alternative path of Eq. (G1) decreases the energy while that with the non-thermal bath along the initial path increases it.

By contrast, our inequality (9) yields the condition (11) and hence the bound (12).

If the Hamiltonian is constant during the energising stroke, then $\tilde{\mathcal{E}}_{d,h} = \Delta E_{\text{pas,h}}|_d + \widetilde{\Delta\mathcal{W}}|_d$, where $\widetilde{\Delta\mathcal{W}}|_d \leq 0$ is the ergotropy lost to the effective thermal bath in the second step of the alternative path in Eq. (G1). A comparison of Eq. (I4) with our bound Eq. (14) for a constant Hamiltonian then yields $\eta_{\text{max}}^{\text{Otto}} \leq \eta_\Sigma$.

If $\mathcal{E}_{d,c} > 0$ ($\Delta E_{\text{pas,h}}|_d < 0$), then also the cold bath provides energy, which has to be taken into account in the efficiency. The latter now reads [29]

$$\eta = \frac{-W}{\mathcal{E}_{d,h} + \mathcal{E}_{d,c}} = \frac{\mathcal{E}_{d,h} + \mathcal{E}_{d,c}}{\mathcal{E}_{d,h} + \mathcal{E}_{d,c}} = 1, \quad (\text{I5})$$

which cannot be further restricted by any inequality for ΔS .

Appendix J: Expressions used in Fig. 8

In Fig. 8 we have used the energies

$$\mathcal{E}_{d,h} = \hbar\omega_h(\bar{n}_h + \Delta\bar{n}_h - \bar{n}_c) \quad (\text{J1a})$$

$$\Delta E_{\text{pas,h}} = \hbar\omega_h(\bar{n}_h - \bar{n}_c) \quad (\text{J1b})$$

$$\tilde{\mathcal{E}}_d = \Delta E_{\text{pas,h}}|_d - \hbar\omega_h\Delta\bar{n}_c. \quad (\text{J1c})$$

Here ω_c (ω_h) is the oscillator frequency before (after) the compression stroke. Furthermore, we have defined $\bar{n}_i = [\exp(\hbar\omega_i/[k_B T_i]) - 1]^{-1}$ and $\Delta\bar{n}_i = (2\bar{n}_i + 1) \sinh^2(r)$ for $i \in \{c, h\}$, where r denotes the squeezing parameter [42]. Additionally, we have used the actual efficiency [29]

$$\eta = 1 - \frac{(\bar{n}_h - \bar{n}_c)\omega_c}{(\bar{n}_h + \Delta\bar{n}_h - \bar{n}_c)\omega_h}. \quad (\text{J2})$$

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